X-ray broadband Ni/SiC multilayers: improvement with W barrier layers

B. Emprin,[1,2,*] Ph. Troussel,[1] G. Soullié,[1] Ph. Stemmler,[1] P. Mercère,[3] E. Meltchakov,[2] A. Jérôme,[2] and F. Delmotte[2]

[1]CEA, DAM, DIF, F-91297 Arpajon, France
[2]Laboratoire Charles Fabry, Institut d'Optique, CNRS, Univ Paris Sud, 2 avenue Augustin Fresnel, 91127 Palaiseau Cedex, France
[3]Synchrotron SOLEIL, L'Orme des Merisiers, Saint-Aubin, BP48, 91192 Gif-sur-Yvette, France
[*]benoit.emprin@cea.fr

Abstract: We present an experimental study and performance improvement of periodic and aperiodic Ni/SiC multilayer coatings. Periodic Ni/SiC multilayer mirrors have been coated and characterized by grazing incidence X-ray reflectometry at 8.048 keV (Cu Kα radiation) and by measurements at 3 keV and 5 keV on synchrotron radiation facilities. An interdiffusion effect is found between Ni and SiC layers. A two-material model, Ni_xSi_y/SiC, using a silicide instead of Ni, was used to fit the measurements. The addition of 0.6 nm W barrier layers at the interfaces allows a significant reduction of the interdiffusion between Ni and SiC. In order to obtain a specific reflectivity profile in the 2 – 8 keV energy range, we have designed and coated aperiodic multilayer mirrors by using Ni/SiC with and without W barrier layers. The experimental reflectivity profiles as a function of the photon energy were measured on a synchrotron radiation facility in both cases. Adding W barrier layers in Ni/SiC multilayers provides a better precision on the layer thicknesses and a very good agreement between the experimental data and the targeted spectral profile.

OCIS codes: (340.7470) X-ray mirrors; (230.4170) Multilayers; (240.0310) Thin films.

References and links

1. F. Schäfers, M. Mertin, D. Abramsohn, A. Gaupp, H.-C. Mertins, and N. Salashchenko, "Cr/sc nanolayers for the water window: improved performance," Nucl. Instrum. Methods Phys. Res. A **467–468**, 349–353 (2001).
2. D. L. Windt, E. M. Gullikson, and C. C. Walton, "Normal-incidence reflectance of optimized W /B_4 C x-ray multilayers in the range 1.4 nm < l < 2.4 nm," Opt. Lett. **27**(24), 2212–2214 (2002).
3. M. Fernández-Perea, M.-A. Descalle, R. Soufli, K. P. Ziock, J. Alameda, S. L. Baker, T. J. McCarville, V. Honkimäki, E. Ziegler, A. C. Jakobsen, F. E. Christensen, and M. J. Pivovaroff, "Physics of reflective optics for the soft gamma-ray photon energy range," Phys. Rev. Lett. **111**(2), 027404 (2013).
4. F. Choueikani, B. Lagarde, F. Delmotte, M. Krumrey, F. Bridou, M. Thomasset, E. Meltchakov, and F. Polack, "High-efficiency B_4C/Mo_2C alternate multilayer grating for monochromators in the photon energy range from 0.7 to 3.4 keV," Opt. Lett. **39**(7), 2141–2144 (2014).
5. A. L. Aquila, F. Salmassi, F. Dollar, Y. Liu, and E. Gullikson, "Developments in realistic design for aperiodic Mo/Si multilayer mirrors," Opt. Express **14**(21), 10073–10078 (2006).
6. C. Bourassin-Bouchet, S. de Rossi, J. Wang, E. Meltchakov, A. Giglia, N. Mahne, S. Nannarone, and F. Delmotte, "Shaping of single-cycle sub-50-attosecond pulses with multilayer mirrors," New J. Phys. **14**(2), 023040 (2012).
7. D. L. Windt, S. Donguy, C. J. Hailey, J. Koglin, V. Honkimaki, E. Ziegler, F. E. Christensen, H. Chen, F. A. Harrison, and W. W. Craig, "W/SiC x-ray multilayers optimized for use above 100 keV," Appl. Opt. **42**(13), 2415–2421 (2003).
8. Z. S. Wang, X. B. Cheng, J. T. Zhu, Q. S. Huang, Z. Zhang, and L. Y. Chen, "Investigation of aperiodic W/C multi-layer mirror for x-ray optics," Thin Solid Films **519**(20), 6712–6715 (2011).
9. F. Bridou, F. Delmotte, P. Troussel, and B. Villette, "Design and fabrication of x-ray non-periodic multilayer mirrors: apodization and shaping of their spectral response," Nucl. Instrum. Methods Phys. Res. A **680**, 69–74 (2012).

10. P. Troussel, B. Villette, B. Emprin, G. Oudot, V. Tassin, F. Bridou, F. Delmotte, and M. Krumrey, "Absolute radiant power measurement for the Au M lines of laser-plasma using a calibrated broadband soft x-ray spectrometer with flat-spectral response," Rev. Sci. Instrum. **85**(1), 013503 (2014).
11. H. Takenaka, H. Ito, T. Haga, and T. Kawamura, "Design and fabrication of highly heat-resistant Mo/Si multilayer soft x-ray mirrors with interleaved barrier layers," J. Synchrotron Radiat. **5**(3), 708–710 (1998).
12. S. Bajt, J. B. Alameda, T. W. Barbee, Jr., W. M. Clift, J. A. Folta, B. Kaufmann, and E. A. Spiller, "Improved reflectance and stability of Mo-Si multilayers," Opt. Eng. **41**(8), 1797–1804 (2002).
13. J. Friedrich, I. Diel, C. Kunz, S. Di Fonzo, B. R. Müller, and W. Jark, "Characterization of sputtered nickel/carbon multilayers with soft-x-ray reflectivity measurements," Appl. Opt. **36**(25), 6329–6334 (1997).
14. H.-C. Mertins, F. Schäfers, H. Grimmer, D. Clemens, P. Böni, and M. Horisberger, "W/C, W/Ti, Ni/Ti, and Ni/V multilayers for the soft-x-ray range: experimental investigation with synchrotron radiation," Appl. Opt. **37**(10), 1873–1882 (1998).
15. F. Eriksson, N. Ghafoor, F. Schäfers, E. M. Gullikson, and J. Birch, "Interface engineering of short-period Ni/V multilayer x-ray mirrors," Thin Solid Films **500**(1–2), 84–95 (2006).
16. S. Bajt, H. N. Chapman, E. Spiller, S. Hau-Riege, J. Alameda, A. J. Nelson, C. C. Walton, B. Kjornrattanawanich, A. Aquila, F. Dollar, E. Gullikson, C. Tarrio, and S. Grantham, "Multilayers for next-generation x-ray sources," Proc. SPIE **6586**, 65860J (2007).
17. H. Höchst, D. W. Niles, G. W. Zajac, T. H. Fleisch, B. C. Johnson, and J. M. Meese, "Electronic-structure and thermal-stability of Ni/SiC(100) interfaces," J. Vac. Sci. Technol. B **6**(4), 1320–1325 (1988).
18. J. H. Gülpen, A. A. Kodentsov, and F. J. J. Vanloo, "Growth of silicides in Ni-Si and Ni-SiC bulk diffusion couples," Z. Metallk. **6**(8), 530–539 (1995).
19. A. S. Edelstein, D. J. Gillespie, S. F. Cheng, J. H. Perepezko, and K. Landry, "Reactions at amorphous SiC/Ni interfaces," J. Appl. Phys. **85**(5), 2636–2641 (1999).
20. C. P. Jensen, K. K. Madsen, and F. E. Christensen, "Investigation of new material combinations for hard x-ray telescope designs," Proc. SPIE **6266**, 626612 (2006).
21. J. Gautier, F. Delmotte, M. Roulliay, F. Bridou, M. F. Ravet, and A. Jérome, "Study of normal incidence of three-component multilayer mirrors in the range 20-40 nm," Appl. Opt. **44**(3), 384–390 (2005).
22. TFCalc, "Thin film design software for windows," Software Spectra Inc., 14025.
23. D. L. Windt, "IMD - software for modeling the optical properties of multilayer films," Comput. Phys. **12**(4), 360–370 (1998).
24. M. Schuster and H. Göbel, "Parallel-beam coupling into channel-cut monochromators using curved graded multilayers," J. Phys. D Appl. Phys. **28**(4A), A270–A275 (1995).
25. M. Idir, P. Mercère, T. Moreno, A. Delmotte, P. Dasilva, M. H. Modi, R. Garrett, I. Gentle, K. Nugent, and S. Wilkins, "Metrology and tests beamline at soleil design and first results," AIP Conf. Proc. **1234**, 485–488 (2010).
26. M. Krumrey and G. Ulm, "High-accuracy detector calibration at the PTB four-crystal monochromator beamline," Nucl. Instrum. Methods Phys. Res. A **467–468**, 1175–1178 (2001).
27. A. Ulyanenkov, "Leptos: a universal software for x-ray reflectivity and diffraction," Adv. Comput. Math. **5536**, 1–15 (2004).
28. M. R. Jackson, R. L. Mehan, A. M. Davis, and E. L. Hall, "Solid-state SiC/Ni alloy reaction," Metall. Mater. Trans., A Phys. Metall. Mater. Sci. **14**(2), 355–364 (1983).
29. R. C. J. Schiepers, F. J. J. Vanloo, and G. Dewith, "Reactions between alpha-silicon carbide ceramic and nickel or iron," J. Am. Ceram. Soc. **71**(6), C284–C287 (1988).
30. A. V. Vinogradov, Y. P. Pershin, E. Zubaryev, D. L. Voronov, O. Penkov, V. V. Kondratenko, Y. A. Uspenskii, I. A. Artioukov, and J. F. Seely, "Structure, thermal stability, and reflectivity of Sc/Si and Sc/W/Si/W multilayer x-ray mirrors," Proc. SPIE **4505**, 230–235 (2001).
31. D. L. Windt, J. A. Bellotti, B. Kjornrattanawanich, and J. F. Seely, "Performance optimization of Si/Gd extreme ultraviolet multilayers," Appl. Opt. **48**(29), 5502–5508 (2009).
32. H. Maury, F. Bridou, P. Troussel, E. Meltchakov, and F. Delmotte, "Design and fabrication of supermirrors for (2-10 keV) high resolution x-ray plasmas diagnostic imaging," Nucl. Instrum. Methods Phys. Res. A **621**(1–3), 242–246 (2010).
33. D. Besnard, "Fusion with the Megajoule laser," J. Phys. Conf. Ser. **112**(1), 012004 (2008).
34. J. L. Bourgade, B. Villette, J. L. Bocher, J. Y. Boutin, S. Chiche, N. Dague, D. Gontier, J. P. Jadaud, B. Savale, R. Wrobel, and R. E. Turner, "DMX: an absolutely calibrated time resolved broadband soft x-ray spectrometer designed for MJ class laser produced plasmas," Rev. Sci. Instrum. **72**(1), 1173–1182 (2001).

1. Introduction

The emergence of new sources and applications in the EUV and X-ray spectral domains raises more stringent requirements on the quality of multilayer mirrors. There is a real need for multilayers with low roughness and well-controlled interfaces for the design of complex components, such as ultra-short-period multilayers [1–3], alternate multilayer gratings [4], EUV broadband mirrors [5] and phase controlled mirrors for attosecond pulses [6], aperiodic broadband mirrors for X-ray optics [7,8] or plasma diagnostics [9,10]. It is well known that interface defects and material chemical stability often limit the theoretical efficiency of such mirrors. Improvement of multilayer stability and/or reduction of interface defects have been achieved for several material combinations by adding a thin layer (so-called "barrier layer", typically 0.3 nm to 1 nm thick) at multilayer interfaces [11,12]. Ni-based multilayers have been investigated in the past for the soft X-ray domain, in particular Ni/C near the K-1s edge of C (at 284 eV), Ni/Ti and Ni/V near the L_3-2p edges of Ti and V (at 454 eV and 512 eV, respectively) [13–15]. Theoretical simulations show that Ni/C is also a good theoretical candidate for grazing X-ray mirrors in the range 2 – 8 keV. Results from Friedrich *et al.* indicate that no intermixing takes place between Ni and C but that significant interface roughness (about 0.6 to 0.7 nm) exists in these multilayers [13]. Thus, we can wonder if other material combinations with smoother interfaces would provide better experimental efficiency. For example, S. Bajt *et al.* have achieved a significant increase in Ni/C peak reflectance near the C edge (at E = 270 eV) by adding a thin B_4C layer at C-on-Ni interfaces [16].

We present here an experimental study of Ni/SiC multilayers for X-ray mirror applications in the range 2 – 8 keV. Previous results reported on Ni/SiC interfaces have been mainly motivated by electronic applications [17–19]. From these studies, we can presume that silicide formation will take place at Ni/SiC interfaces. Jensen *et al.* have also reported some interesting results on $Ni_{0.93}V_{0.07}$/SiC multilayers for hard X-ray telescope applications. They have studied the evolution of interface roughness as a function of the multilayer d-spacing and they have found that the interface roughness is too high to produce efficient small d-spacing mirrors required for such applications [20]. In this paper, we investigate the effect of adding barrier layers in Ni/SiC coatings. We have used grazing incidence X-ray reflectometry at different photon energies in order to characterize and model the interfaces in periodic Ni/SiC multilayers with and without W barrier layers. We will present the development of aperiodic broadband mirrors in the 4 – 6 keV range, based on Ni/SiC multilayers and compare the experimental results with and without W barrier layers. Finally, we will discuss the interest of using W barrier layers in Ni/SiC multilayer systems for the design of aperiodic mirrors.

2. Experimental tools

The mirrors were coated by magnetron sputtering at the "Institut d'Optique", using a MP800S apparatus from Plassys®, especially designed to realize multilayer mirrors with thin thicknesses [21]. The sputtering gas was argon whose pressure was fixed at 2 mTorr (0.093 Pa) in the deposition chamber. The plasma discharge was established with an RF power of 65 W for the Ni target and 150 W for the SiC target. We used a DC current of 70 mA for the W target. The coatings were deposited on silica superpolished substrates from Winlight Optics®, with 25.4 mm diameter and surface roughness less than 0.3 nm rms in the mid- and high-spatial frequency ranges. To optimize the aperiodic multilayer mirror reflectivity, we used a commercial code: TFCalc®, with the needle procedure [22]. This code was developed first for the visible light domain, and we imported the optical constants for X-ray wavelengths. Nevertheless, this code does not take into account the interface roughness. To determine the calculated spectral profile with the roughness included, we used the IMD simulation software [23] that is a free point-and-click IDL application to calculate reflectivity of multilayer structures. Uncertainties of thicknesses have been estimated by systematic variation of each parameter independently (i.e. for each material).

Grazing incidence X-ray reflectometry (GIXR) was made with a commercial grazing incidence reflectometer (BRUKER® Discover D8) working with Cu Kα radiation at energy E = 8.048 keV (wavelength λ = 0.154 nm). A collimating Göbel mirror [24], located at the exit of the X-ray tube, provides high flux density and low divergence beam. A rotary absorber, combined with a scintillator, provides a linear detection over 7 decades. Soller slits and 0.1 mm divergence slits are used on the incident and reflected beams. The reflectivity curve is obtained by varying the grazing incidence angle while tracking the reflected beam (θ-2θ scan configuration). The mechanical angular accuracy and angular resolution are better than 0.01°. Reflectivity measurements were also realized on the Metrology and Tests beamline at the synchrotron radiation facility SOLEIL [25]. Built on a bending magnet, this beamline is composed of two branches covering an energy range between 30 eV and 40 keV and offering also access to the white-light beam from the source. Experiments were performed at 3 and 5 keV on the hard X-ray branch using the Si(111) Double-Crystal Monochromator. The mirrors were placed in an ultra-high vacuum (UHV) reflectometer, which is specified to have an angular resolution of 0.0005°, with an accuracy of 0.0003°, for both the sample and detector rotation stages. The configuration of our experiments permitted an angular resolution of about 0.005°. The beam size used for these experiments was 0.15 mm vertically and 2.0 mm horizontally. During our measurements, we used a silicon photodiode Hamamatsu K6517B, with different apertures mounted on the detector arm. The intensity was measured by a 6517B KEITHLEY amperemeter.

Experiments were also performed at the four-crystal monochromator beamline in the Physikalisch-Technische Bundesanstalt (PTB) laboratory at the synchrotron radiation facility Bessy II [26]. The energy range of this line is 1.75 – 10 keV using either Si or InSb crystals in the monochromator. The mirrors were placed in a UHV reflectometer, which provides a 0.001° angular resolution for the sample and the detector. The typical beam size is 0.3 mm vertically and 0.5 mm horizontally. The vertical beam divergence is about 0.02°. Silicon photodiodes with different apertures and a photon counting detector are mounted on the detector arm. To account variations in the incident monochromatic photon flux, the currents are normalized to the current of a thin photodiode operating in transmission in front of the reflectometer. We analyzed the reflectivity data with the program Leptos® [27] that permits to fit the GIXR and SOLEIL data using a genetic algorithm. The fit of the measured reflectivity curve gives the parameters of the layer thicknesses, materials density and the average interface roughness.

3. Realization of periodic multilayer mirrors

3.1. Ni/SiC periodic multilayer mirrors

We have coated and characterized four Ni/SiC periodic multilayer samples. The samples named A, C and D are composed of 15 periods of Ni/SiC: the stack formula is 15[SiC/Ni]/silica substrate. Sample B has a similar stack formula with 30 periods instead of 15. The expected thicknesses of the four samples, estimated from the Ni and SiC deposition rates, are presented in Table 1. The deposition rates of Ni and SiC materials were calculated from GIXR analysis of single layer samples. We characterized each sample successively by GIXR measurements at 8.048 keV at the Charles Fabry Laboratory (LCF) and by reflectometry measurements at 5 and 3 keV at SOLEIL. Each reflectivity spectrum has been fitted independently by using a model with two materials (Ni and SiC) in each period [Fig. 1(a)]. Figure 2 displays an example of experimental and fitted reflectivity spectra for sample A at 8.048 keV, 5 keV and 3 keV. The fitted parameters in terms of thickness and density of each material are given in Table 1. For all fits, the Ni-on-SiC and SiC-on-Ni interface roughnesses are in the range 0.15-0.35 nm and 0.20-0.40 nm, respectively. Notice that these values are significantly lower than the roughness values reported previously for $Ni_{0.93}V_{0.07}$/SiC multilayers (in the range 0.40 – 0.60 nm for equivalent d-spacing) [20]. Overall, we obtain a good agreement between thickness values fitted at 3 keV, 5 keV and 8.048 keV. Variations of the layer thickness around the average result for each sample are within the fit uncertainties (estimated to be about ± 0.05 nm for each layer thickness). We can notice that the Ni and SiC thicknesses are respectively higher and lower than expected. Such behaviour is generally attributed to interdiffusion between the two materials. For sample A, B and C, the Ni thickness is approximately 1.00 to 1.10 nm higher than expected. For sample D, the Ni thickness is only 0.70 nm higher than expected. The main difference between samples

A, B and C with sample D is that the targeted SiC thickness is not the same. The expected SiC thickness in sample D is half that of the other samples.

Table 1. Fitted values of the Ni/SiC periodic multilayer mirrors. They were obtained by the Leptos software after measurements on the Metrology and Tests beamline at the SOLEIL synchrotron at 3 and 5 keV and at LCF at 8.048 keV. The curves of sample A using the fitted values presented here are shown in Fig. 2. The uncertainties of the fit results are ± 0.05 nm for Ni and SiC. The uncertainties of the fitted densities are ± 0.1 g/cm^3.

Mirror	Number of periods	Material	Targeted thickness (nm)	Fitted thickness (nm)			Fitted density (g.cm^{-3})		
				3 keV	5 keV	8 keV	3 keV	5 keV	8 keV
A	15	Ni	1.70	2.65	2.65	2.63	6.5	6.9	7.2
		SiC	3.82	2.75	2.75	2.77		3.22	
B	30	Ni	2.27	3.41	3.40	3.39	6.7	6.8	7.3
		SiC	3.83	2.50	2.51	2.52		3.22	
C	15	Ni	3.40	4.43	4.53	4.58	7.3	7.5	7.8
		SiC	3.82	2.64	2.54	2.47		3.22	
D	15	Ni	1.70	2.32	2.41	2.43	7.0	7.3	7.7
		SiC	1.91	1.19	1.10	1.08		3.22	

Table 2. Fitted values of density for samples A, B and C with the Ni_5Si_2/SiC model. They were obtained by the Leptos software. The uncertainties of the fit results are ± 0.1 g/cm^3.

Mirror	Composition (nm)	Material	Fitted density (g.cm^{-3})		
			3 keV	5 keV	8 keV
A	15 (Ni 1.70/SiC 3.82)	Ni_5Si_2	6.7	6.8	6.9
		SiC		3.22	
B	30 (Ni 2.27/SiC 3.83)	Ni_5Si_2	6.8	6.7	7.0
		SiC		3.22	
C	15 (Ni 3.40/SiC 3.82)	Ni_5Si_2	7.6	7.6	7.5
		SiC		3.22	

Thus, it seems that the SiC thickness has an impact on the interdiffusion effect between Ni and SiC layers. Thicker SiC layers lead to an increase of interdiffusion. Moreover, in order to fit the experimental critical angle, we had to decrease the average density of materials in the model. Because Ni is the heavy material in the coating, we chose to fit only the Ni density. For each sample, and at each energy, we fitted a Ni density lower (72% to 89%) than the theoretical one (8.902 g.cm^{-3}). It appears that we cannot fit the critical angle at different photon energies with the same value of Ni density. This suggests that the Ni layer in our model is not composed of pure Ni in reality. These results on density variation and on thickness variation indicate the formation of an intermixed layer (most likely a nickel silicide layer).

The interface reactions of Ni/SiC have been investigated by several researchers [17–19] [28,29]. They identified that the reaction products were Ni_2Si, Ni_5Si_2, Ni_3Si and C. In general, the solubility of C in the silicides is limited. Due to the high reactivity of Ni with Si, we suppose that the interdiffusion occupies the entire Ni layer. Thus, we propose to use a model with Ni_xSi_y/SiC instead of Ni layers [Fig. 1(b)]. Under this hypothesis, we obtained realistic fits for samples A, B and C with a Ni_5Si_2/SiC multilayer stack. We kept the layer thicknesses fixed to the previous values and we fitted the Ni_5Si_2 density. The results are presented in Table 2. The Ni_5Si_2-on-SiC and SiC-on-Ni_5Si_2 interface roughnesses are fitted in the range 0.15 – 0.30 nm and 0.20 – 0.35 nm, respectively. With this model, we obtained a fitted

#216865 - $15.00 USD Received 11 Jul 2014; accepted 10 Sep 2014; published 14 Oct 2014

density for the Ni_5Si_2 layers that is almost identical for the 3 keV, 5 keV and 8.048 keV measurements. Nevertheless, Table 2 shows that some small variations of the fitted Ni_5Si_2 density exist between the three samples. This confirms that the interdiffusion effect between Ni and SiC is dependent on the layer thickness. A more complex model with three or four layers in each period (Ni, SiC and Ni_xSi_y and one or both interfaces) would probably be more efficient to fit these phenomena. However, it would raise difficulties in the design of aperiodic multilayers so we decided to use the two material model for this application (see part 4).

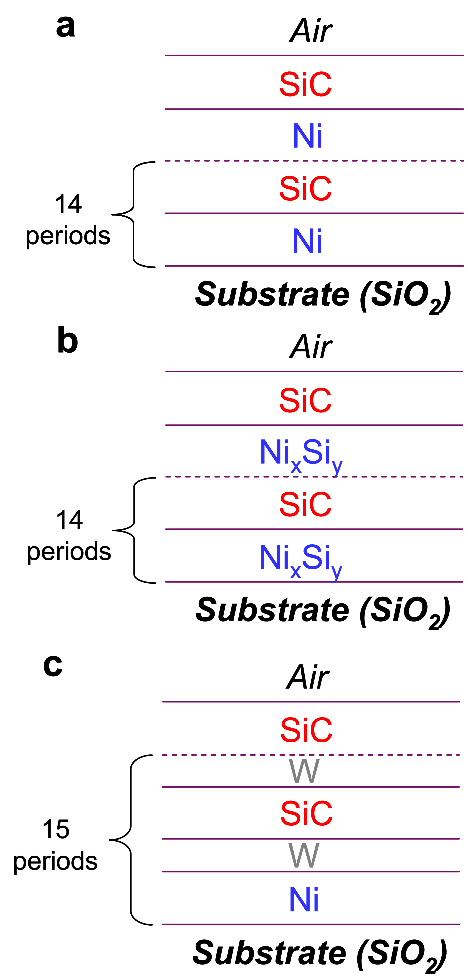

Fig. 1. Periodic multilayer structures: (a) consisting of 15 Ni/SiC periods. (b) consisting of 15 Ni_XSi_Y/SiC periods with the silicide model. (c) consisting of 15 Ni/W/SiC/W periods and a top layer of SiC to protect the stack against oxidation .

#216865 - $15.00 USD Received 11 Jul 2014; accepted 10 Sep 2014; published 14 Oct 2014

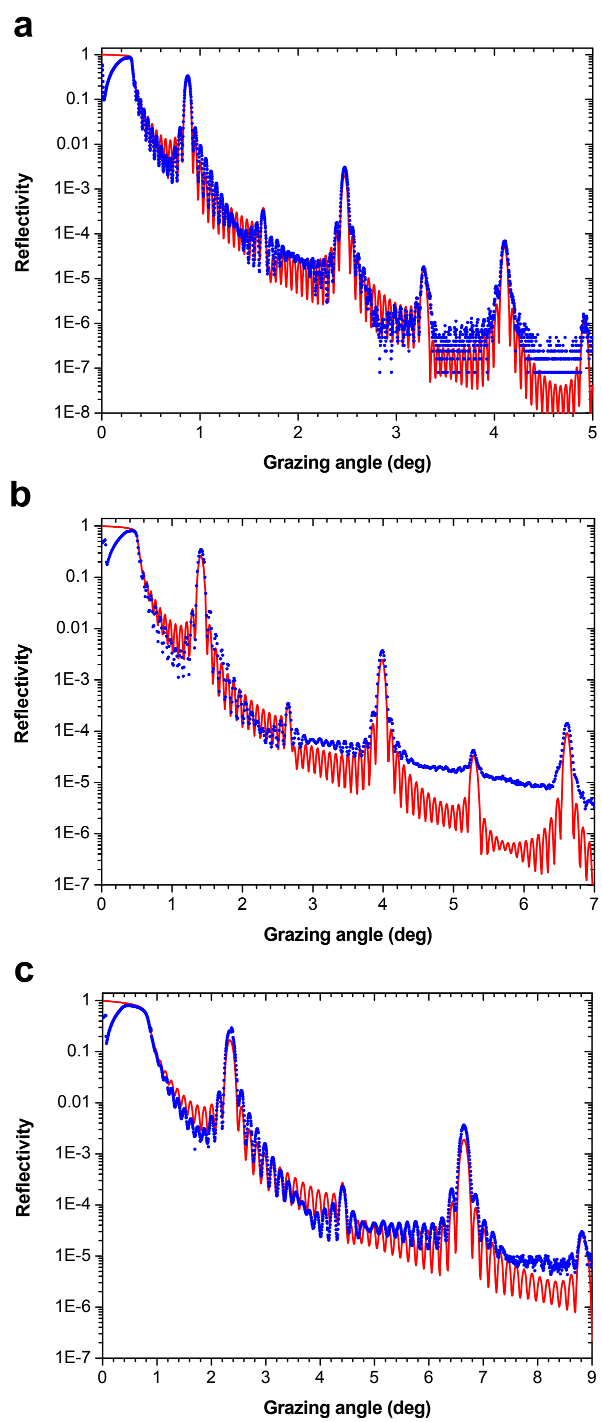

Fig. 2. Reflectivity measurements (dots) of sample A: (a) at E = 8.048 keV at LCF; (b) at E = 5 keV and (c) at E = 3 keV on the Metrology and Tests beamline at SOLEIL. For the LCF and SOLEIL measurements, the fits (solid line) were obtained by the Leptos software. All fitted values are presented in Table 1.

#216865 - $15.00 USD Received 11 Jul 2014; accepted 10 Sep 2014; published 14 Oct 2014

3.2. Ni/W/SiC/W periodic multilayer mirrors

In order to limit the interdiffusion phenomena between the Ni and SiC layers, we decided to add a thin layer of W at both interfaces. It has been previously demonstrated that the use of W at barrier layers in Si-based multilayer as Sc/Si [30] or Si/Gd multilayers [31], significantly reduces the silicide formation at interfaces. We chose to fix the thickness of these W layers to 0.60 nm. This value seems to be a good compromise to limit the interdiffusion and the absorption in the W layers [31]. We coated four new periodic multilayer samples with W barrier layers (samples E, F, G and H). The expected thicknesses for these four samples are given in Table 3. We added a 3.00 nm SiC layer at the top of the stack, as shown in Fig. 1(c), because it presents a better stability to atmosphere than W. The corresponding stack formula for samples E, F and G is SiC/15[W/SiC/W/Ni]/silica substrate. For these three samples, we kept the expected thickness of Ni and SiC the same as for samples A, C and D, respectively. For the sample H, with 30 periods, we decreased the expected thickness of Ni and SiC layers by 0.6 nm in order to keep the total expected period thickness the same as for sample B. All samples were measured by GIXR at 8.048 keV. Sample H was also measured at 5 keV at SOLEIL. Figure 3 displays the reflectivity spectra of this mirror measured at 8.048 keV (a) and at 5 keV (b). In order to limit the number of fitting parameters, we decided to fix the thickness of W layers to its expected value (0.60 nm). In this model, we did not take into account the intermixed regions between Ni-W or SiC-W interfaces. However, this model provides good results towards estimating the layer thicknesses. One can see in Table 3 that with this model, we found the Ni thicknesses approximately equal to the targeted ones.

Table 3. Fitted values of the Ni/W/SiC/W periodic multilayer mirrors. They were obtained by the Leptos software after measurements at LCF at 8.048 keV. The W barrier layers are fixed at 0.60 nm. The sample H was also measured on the Metrology and Tests beamline at SOLEIL at 5 keV. The curves of sample H using the fitted values presented here are shown in Fig. 3. The uncertainties of the thickness fit results are ± 0.05 nm for Ni and SiC.

Mirror	Number of periods	Material	Targeted thickness (nm)	Fitted thickness (nm) 8 keV	Theoretical density ($g.cm^{-3}$) 8 keV
E	15	Ni	1.70	1.78	8.90
		SiC	3.82	3.12	3.22
		SiC (top layer)	3.00	3.47	3.22
F	15	Ni	3.40	3.49	8.90
		SiC	3.82	3.13	3.22
		SiC (top layer)	3.00	3.43	3.22
G	15	Ni	1.70	1.78	8.90
		SiC	1.91	1.30	3.22
		SiC (top layer)	3.00	3.52	3.22
H	30	Ni	1.67	1.67	8.90
		SiC	3.23	2.82	3.22
		SiC (top layer)	3.00	3.50	3.22

The fitted SiC thicknesses are lower than expected with a difference ranging from 0.4 to 0.7 nm. This contraction of SiC thickness has already been observed on W/SiC coatings [32]. The SiC top layer was fitted separately from the other SiC layers because of the ambient atmosphere effects. We found an increase of the SiC layer thickness (about 3.50 nm instead of 3.00 nm). This is most likely attributed to the formation of silicon oxide on top of the SiC layer. The Ni-on-W, W-on-Ni, SiC-on-W and W-on-SiC interface roughnesses are fitted in the range 0.25 – 0.40 nm, 0.25 – 0.35 nm, 0.30 – 0.45 nm and 0.20 – 0.30 nm, respectively. All densities used for fits correspond to the theoretical density of materials. We compare in Fig. 4 the first Bragg peak measured at 5 keV on the two samples with 30 periods (sample B without W, and sample H with W layers). We can see that, even if the W layers are efficient towards reducing the interdiffusion between Ni and SiC, the Bragg peak reflectivity is higher for sample B without the W barrier layers. This is mainly due to the high absorption of W at this energy. One can think to decrease the W barrier layer thickness in order to reduce the absorption. However, simulations with 0.3 nm W layer thickness show that the reflectivity is still lower than the one of Ni/SiC without W barrier. Thus, the choice to fix the thickness value of W layers to 0.60 nm seems to be a good compromise.

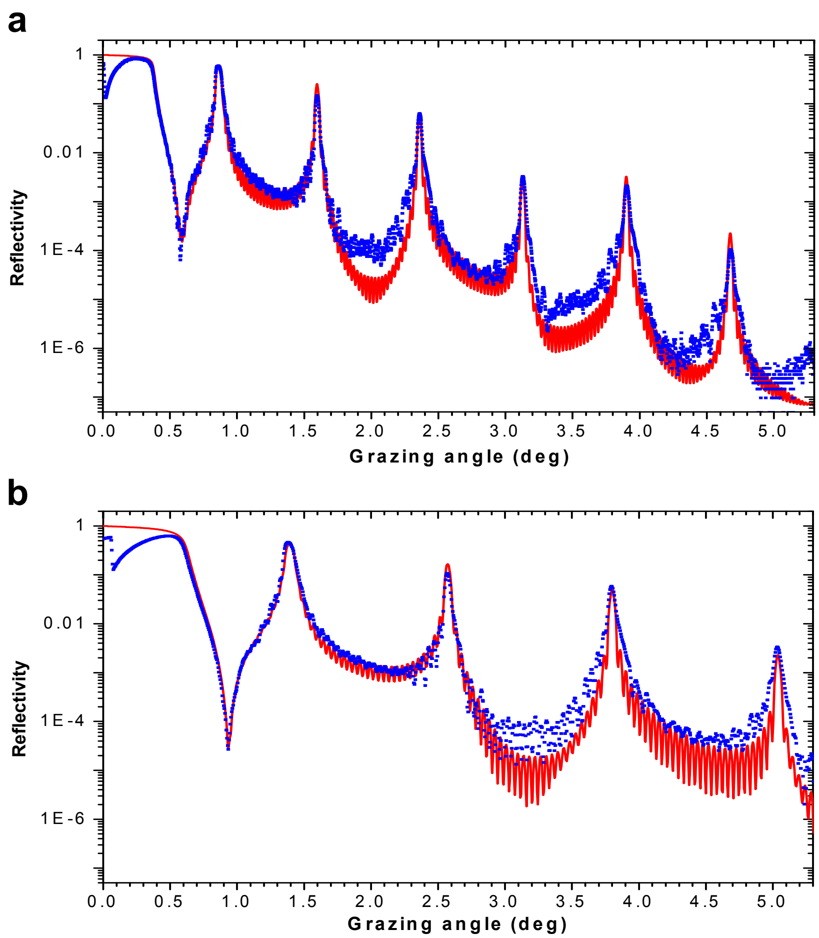

Fig. 3. Reflectivity measurements (dots) of sample H: (a) at E = 8.048 keV at LCF and (b) at E = 5 keV on the Metrology and Tests beamline at SOLEIL. For the LCF measurements, the fits (solid line) were obtained by the Leptos software. Their fitted values are presented in Table 3. For the SOLEIL measurements, the fitted parameters given in Table 3 were used to simulate the 5 keV reflectivity spectrum (solid line).

#216865 - $15.00 USD Received 11 Jul 2014; accepted 10 Sep 2014; published 14 Oct 2014

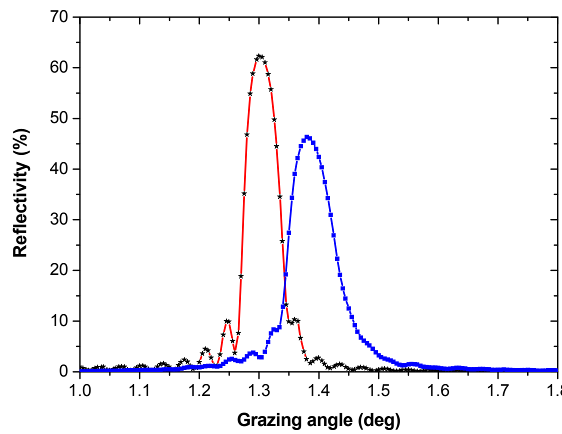

Fig. 4. Reflectivity measurements of the first Bragg peak for samples B (star dots with red line) and H (square dots with blue line). They were obtained at 5 keV on the Metrology and Tests beamline at SOLEIL.

4. Development of aperiodic multilayer mirrors

4.1. Context

Within the framework of its research on Inertial Confinement Fusion (ICF), the "Commissariat à l'énergie atomique et aux énergies alternatives" (CEA) studies and designs advanced X-ray diagnostics in order to probe dense plasmas that will be produced in the future Laser MegaJoule (LMJ) facility [33]. Particularly, CEA implemented an absolutely calibrated broadband soft X-ray spectrometer called DMX [34]. A new channel of this spectrometer is developed for the 4 – 6 keV range and follows another study already made for the 2 – 4 keV channel [9,10].This channel is composed of a 10 µm Fe filter, a multilayer mirror (MIM) and a photoelectric coaxial detector. The MIM is optimized to be used at 1.3° grazing incidence angle. For our study, the shape of the MIM-requested reflectivity is imposed by the spectral response of other channel components. The average reflectivity in the energy band 4 – 6 keV has to be maximized and the reflectivity outside this band has to be as low as possible. We used a 10 µm Fe filter to suppress the total reflection that occurs on the MIM for energies lower than 3 keV.

4.2. Aperiodic Ni/SiC multilayer

In section 3.1., we reveal the existence of an important interdiffusion phenomenon between Ni and SiC layers. Our results suggest that a simple model, such as a Ni_5Si_2/SiC structure, allows fitting the experimental data with a good precision for periodic multilayer samples with different values of layer thickness. Using the IMD software, we generated optical refractive indexes for the Ni_5Si_2 chemical compound with a density fixed at 7.00 g/cm^3. We then optimized a Ni_5Si_2/SiC aperiodic multilayer mirror with a reflectivity target spectrum plotted as solid line in Fig. 5. The simulated reflectivity of the optimized MIM is plotted as dash line in Fig. 5. The MIM consists of 80 layers with thickness ranging from 2.00 to 4.15 nm. The reflectivity measurement of this sample was performed at PTB under 1.3° grazing incidence angle between 2.5 keV and 8 keV [Fig. 5]. We can see that the experimental reflectivity of the mirror is not in good agreement with the calculated one. There are some strong mismatches in the reflectivity profile in the 4 – 6 keV energy band, the bandwidth is not reproduced and there is a shift of about 200 eV towards low energies. This shift indicates that the average thickness of the layers in the stack is higher than expected. This first attempt, to realize an aperiodic mirror with Ni/SiC multilayer, shows that the simple Ni_5Si_2/SiC model is not sufficient. Aquila *et al.* demonstrated the need to include interdiffusion in the development and optimization of aperiodic Mo/Si multilayer structures in the EUV [5]. The need for high precision on each layer thickness in such a coating would require probably an even more complex model to take into account the variations of interdiffusion effects between Ni and SiC as a function of layer thickness.

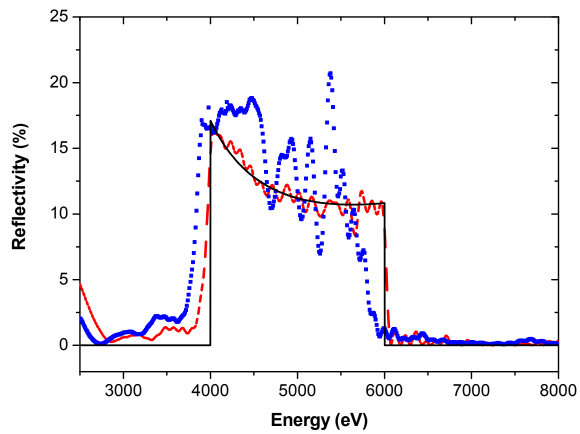

Fig. 5. Reflectivity measurements obtained with the first Ni/SiC (Ni_5Si_2/SiC) aperiodic stack (N = 80 layers): Shown are the targeted (solid line), calculated (dash line) reflectivity profiles, and the measured reflectivity profile obtained at PTB (dots), under 1.3° grazing incidence angle between 2.5 keV and 8.0 keV including the nominal bandwidth (4 – 6 keV).

4.3. Aperiodic Ni/W/SiC/W multilayer

We optimized another aperiodic MIM with 0.6 nm W barrier layers by using the model presented in section 3.2. The stack formula was optimized taking into account a 0.40 nm average interface roughness. The optimized MIM is composed of 82 layers in Ni/W/SiC/W sequences. Layer thickness ranges from 1.20 nm to 5.38 nm (and 0.60 nm for the W layers). A top layer of SiC was added as capping layer and its thickness was optimized to 6.78 nm. After deposition and GIXR measurement at 8.048 keV, we can see in Fig. 6(a) that the measured reflectivity is in very good agreement with the calculated one. In order to confirm this result, we measured this mirror at SOLEIL under 1.3° grazing incidence in the 3 – 8 keV energy band [Fig. 6(b)]. As expected from the angular measurement, the measured reflectivity is really close to the calculated formula. Indeed, the reflectivity outside the 4 – 6 keV energy band is very low ($R_{max} \leq 1.6\%$). Some small oscillations remain (between 5.2 keV and 5.9 keV) due to the sum of small differences between the real and expected thicknesses of each layer in the stack, but the overall spectral profile is in very good agreement within the targeted one. These results clearly show that the use of W barrier layers in Ni/SiC coatings is very beneficial for aperiodic mirror applications. Although the Bragg peak reflectivity for periodic Ni/SiC coatings is reduced by the addition of W layers, we were able to design a broadband mirror with the same average in-band reflectivity with or without W barriers (see dash lines in Figs. 5 and 6). The good agreement between simulated and experimental results shown in Fig. 6 demonstrates that, by using W barrier layers, we have achieved a very high accuracy on each layer thickness. Moreover, the addition of W layers in the design allows reducing significantly the number of Ni and SiC layers. Indeed, the Ni/SiC aperiodic mirror is composed of 40 Ni and 40 SiC layers with variable thickness whereas the Ni/W/SiC/W one is composed of 21 Ni and 21 SiC layers with variable thickness and 41 W layers with fixed thickness. Thus, the experimental development is facilitated by the use of W barrier layers.

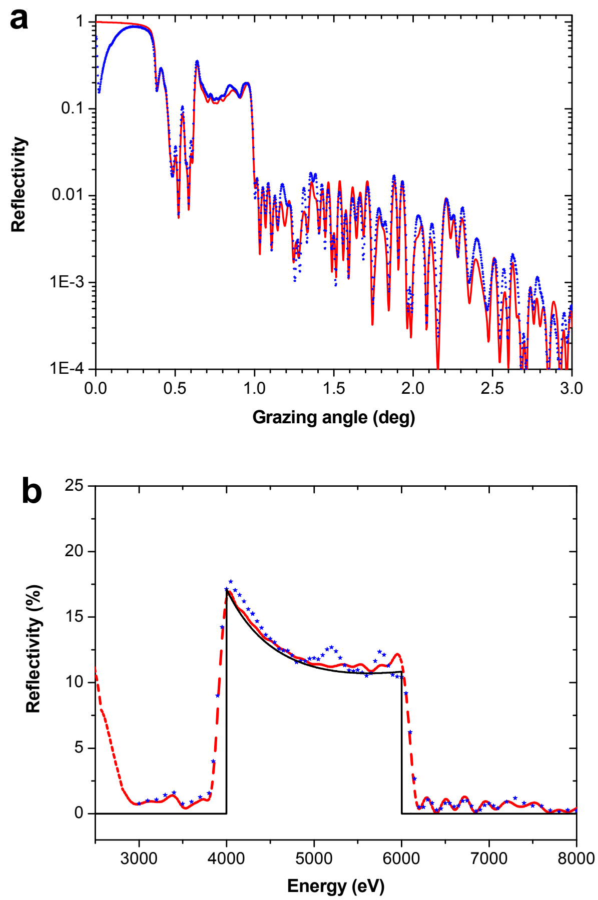

Fig. 6. Reflectivity measurements obtained with the Ni/W/SiC/W aperiodic multilayer (N = 83 layers): (a) GIXR measurements at E = 8.048 keV (dots) are compared with the expected reflectivity design (solid line) ; (b) SOLEIL measurements under 1.3° grazing incidence angle (dots), calculated (dash line) and targeted (solid line) reflectivity profiles.

5. Conclusions

Periodic Ni/SiC multilayer mirrors have been developed and characterized by GIXR at 8.048 keV and by reflectometry measurements at 3 keV and 5 keV on a synchrotron radiation source. We have shown that significant interdiffusion takes place between Ni and SiC layers. We demonstrated that reflectivity measurements can be fitted by using a simple Ni_5Si_2/SiC model. We have also studied Ni/SiC periodic multilayers with 0.6 nm W barrier layers at each interface. Our results show that the W barrier layers efficiently reduce the interdiffusion between Ni and SiC. However, the reflectivity of Ni/SiC periodic multilayers without W barrier layers is somewhat higher than with W barrier layers. Aperiodic multilayer mirrors were optimized by using Ni/SiC with and without barrier layers in order to achieve a specific reflectivity profile in the 4 – 6 keV energy band and an out-of-band reflectivity as low as possible. In both cases, we were able to optimize a theoretical formula with a spectral response close to the targeted one. For Ni/SiC without W barrier layers, the interdiffusion between Ni and SiC prevents us from obtaining a sufficient precision on the thickness of each layer in the stack and the experimental reflectivity profile is not in good agreement with the targeted one. By adding W barrier layers, we achieved a better precision on the layer thicknesses and a very good agreement between the experimental and the targeted spectral profile.

Following this study, an aperiodic Ni/W/SiC/W coating has been deposited on a silica superpolished substrate with dimensions 90 x 25 x 10 mm^3. This mirror will be implemented on the new 4 – 6 keV channel of the DMX spectrometer on hohlraum experiments, at the OMEGA laser facility of Laboratory for Laser Energetics from University of Rochester (USA). This kind of aperiodic mirror, able to select broadband bandwidth typically with $\Delta E/E = 2.5$, can be used for spectral filtering in other X-ray instruments, for example X-ray imaging diagnostic. As already reported by D. Windt, Ni/SiC multilayer mirrors are also a good candidate towards high reflectivity mirrors at higher energies for hard X-ray applications [7,20].

Acknowledgments

All multilayer depositions have been carried out on the deposition machine of CEMOX (Centrale d'Élaboration et de Métrologie d'Optique X), a platform of LUMAT federation (CNRS FR2764). The authors would like to thank SOLEIL beamline staff and CEA associates (project submitted to peer review committee: No. 20130059) for measurements on the hard X-ray branch of the Metrology and Tests beamline. The authors would like also to thank Michael Krumrey and the PTB beamline staff for measurements on the four-crystal monochromator beamline. The authors would like to thank Regina Soufli from Lawrence Livermore National Lab (USA) for fruitful discussions.